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#### (54) [Title of the Invention]

# Resin for Use in Sheets, and Resin Sheet Obtained Using Said Resin

#### (57) [Summary]

[Object] To provide a calenderable noncrystalline polyester resin whose hardness can be freely adjusted by varying the amount of the introduced additive, and which has good elongation at low temperatures, and to provide a resin sheet obtained using the resin.

[Means of Achievement] A glycerol fatty acid ester, or a polymer or polyether containing glycol as a constituent unit is added to a noncrystalline polyester resin.

#### [Claims]

[Claim 1] A resin for use in sheets, obtained by adding a glycerol fatty acid ester, or a polymer or polyether containing glycol as a constituent unit to a noncrystalline polyester resin.

[Claim 2] The resin for use in sheets according to 1, wherein the polymer is polyethylene glycol or polypropylene glycol.

[Claim 3] The resin for use in sheets according to 1, wherein the polyether is polyethylene glycol monomethyl ether or polyethylene glycol dimethyl ether.

[Claim 4] The resin for use in sheets according to 1, wherein the glycerol fatty acid ester is glycerol diacetomonolaurate or glycerol diacetooleate.

[Claim 5] A resin sheet composed of a noncrystalline polyester resin obtained by the addition of a glycerol fatty acid ester, or a polymer or polyether containing glycol as a constituent unit.

[Claim 6] The resin sheet according to 5, wherein the polymer is polyethylene glycol or polypropylene glycol.

[Claim 7] The resin sheet according to 5, wherein the polyether is polyethylene glycol monomethyl ether or polyethylene glycol dimethyl ether.

[Claim 8] The resin sheet according to 5, wherein the glycerol fatty acid ester is glycerol diacetomonolaurate or glycerol diacetooleate.

### [Detailed Description of the Invention]

[0001]

[Technological Field of the Invention] The present invention relates to a resin for use in sheets, and a resin sheet obtained using this resin.

#### [0002]

[Prior Art] A demand for resins that can be used as substitutes for polyvinyl chloride (PVC) resins has recently arisen due to environmental concerns. Examples of substitutes for hard vinyl chloride include polyethylene terephthalate (PET) and polystyrene (PS), whereas examples of substitutes for soft vinyl chloride include polypropylene (PP), polyethylene (PE), ethylene vinyl acetate (EVA), ethylene methyl acrylate (EMA), and ethylene methyl methacrylate (EMMA). Blends thereof have also been proposed.

#### [0003]

[Problems to Be Solved by the Invention] With conventional vinyl chloride resin substitutes, however, the resin hardness is determined by the resin type and is difficult to vary for sheet manufacturers, who calender, extrude, and otherwise process such resins. In addition,

pretreatments such as corona treatments and plasma treatments are often required in the case of fabrication processes such as printing and bonding.

[0004] An object of the present invention is to provide a resin for use in sheets whose properties or behavior traits are similar to those of a soft or semihard vinyl chloride resin, which can be calendered, whose hardness can be freely adjusted by varying the amount of the introduced additive, and which have good elongation at low temperatures (for example, 23°C to 40°C), and to provide a sheet composed of this resin.

#### [0005]

[Means Used to Solve the Above-Mentioned Problems] The inventors perfected the present invention upon discovering that a resin which is suitable for use in soft or semihard sheets and in which the characteristics of a noncrystalline polyester resin capable of being printed, welded, or otherwise processed are utilized can be obtained by adding a glycerol fatty acid ester, or a polymer or polyether containing glycol as a constituent unit to a noncrystalline polyester resin; and that the hardness can be adjusted by varying the amount of the introduced additive.

[0006] The resin for use in sheets according to the present invention is characterized in being obtained by adding a glycerol fatty acid ester, or a polymer or polyether containing glycol as a constituent unit to a noncrystalline polyester resin. As used herein, the term "sheet" refers to a soft or semihard sheet, and this term also includes films.

[0007] The resin sheet of the present invention is characterized in being composed of a noncrystalline polyester resin obtained by the addition of a glycerol fatty acid ester, or a polymer or polyether containing glycol as a constituent unit.

[0008] The noncrystalline polyester resin is a resin obtained by the polycondensation of adipic acid, terephthalic acid, isophthalic acid, or another dicarboxylic acid; ethylene glycol; and 1,4-cyclohexanediol, 1,3-propanediol, 1,4-butanediol, or another diol. The amount of 1,4-cyclohexanediol residue is preferably greater than the amount of ethylene glycol residue because of calendering considerations.

[0009] The glycol may be ethylene glycol or propylene glycol. The polymer may be polyethylene glycol or polypropylene glycol, and the polyether may be polyethylene glycol monomethyl ether, or polyethylene glycol dimethyl ether. The number-average molecular weight of the polymer or polyether is preferably 300 to 20,000.

JP 2002 – 294043 A Page 3

[0010] The glycerol fatty acid ester may be glycerol diacetomonolaurate or glycerol diacetooleate.

[0011] As an additive, the glycerol fatty acid ester, or the polymer or polyether containing glycol as a constituent unit is preferably added in an amount of 5 to 30 parts per 100 parts of noncrystalline polyester resin. Adding less than 5 parts fails to produce a softening effect, whereas adding more than 30 parts results in an excessively reduced melt viscosity, and hence impairs sheeting, and tends to force the additive to the surface if a sheet is formed.

[0012] Polyethylene wax or another lubricant may also be added as needed.

[0013] The components are melted in a Banbury mixer and are formed into a sheet with a thickness of, for example, 80 to 800  $\mu$ m by using a calendaring machine.

#### [0014]

#### [Working Examples]

[0015] < Working Example 1 and 2> Polyethylene glycol dimethyl ether (molecular weight: 500) and a lubricant were added in the ratios shown in Table 1 to 100 parts of noncrystalline polyester resin (Eastar PET-G 6763, manufactured by Eastman Chemical Japan), melted in a Banbury mixer, and formed into a sheet with a thickness of 200 µm by using a calendering machine. Polyethylene wax was used as the lubricant.

#### [0016]

#### [Table 1]

	Noncrystalline polyester resin	Polyethylene glycol dimethyl ether	Lubricant
Working Example 1	100 parts	10 parts	l part
Working Example 2	100 parts	20 parts	1 part

[0017] < Working Examples 3 and 4> Glycerol diacetate monolaurate and a lubricant (the same as described in Working Example 1 and 2) were added in the ratios shown in Table 2 to 100 parts of noncrystalline polyester resin (the same as described in Working Example 1 and 2), melted in a Banbury mixer, and formed into a sheet with a thickness of 200 µm by using a calendering machine.

#### [0018]

#### [Table 2]

	Noncrystalline polyester resin	Glycerol diacetomonolaurate	Lubricant
Working Example 3	100 parts	10 parts	1 part
Working Example 4	100 parts	20 parts	1 part

[0019] < Comparative Examples 1 to 3> Polyethylene glycol dimethyl ether (molecular weight: 500) and a lubricant (the same as described in Working Example 1 and 2) were added in the ratios shown in Table 3 to 100 parts of noncrystalline polyester resin (the same as described in Working Example 1 and 2), melted in a Banbury mixer, and formed into a sheet with a thickness of 200 µm by using a calendering machine.

#### [0020]

#### [Table 3]

	Noncrystalline polyester resin	Polyethylene glycol dimethyl ether	Lubricant
Comparative Example 1	100 parts	_	1 part
Comparative Example 2	100 parts	3 parts	1 part
Comparative Example 3	100 parts	35 parts	l part

[0021] < Comparative Examples 4 and 5> Glycerol diacetate monolaurate and a lubricant (the same as described in Working Example 1 and 2) were added in the ratios shown in Table 4 to 100 parts of noncrystalline polyester resin (the same as described in Working Example 1 and 2), melted in a Banbury mixer, and formed into a sheet with a thickness of 200 µm by using a calendering machine.

#### [0022]

#### [Table 4]

	Noncrystalline polyester resin	Glycerol diacetomonolaurate	Lubricant
Comparative Example 4	100 parts	3 parts	1 part
Comparative Example 5	100 parts	35 parts	l part

[0023] < Comparative Examples 6 to 9> A reinforcing agent (MBS), DOP (diethyl hexyl phthalate<sup>1</sup>), and a stabilizer were added in the ratios shown in Table 5 to a polyvinyl chloride

JP 2002 – 294043 A Page 5

<sup>&</sup>lt;sup>1</sup> Translator's note: Diethyl hexyl phthalate is usually abbreviated as "DEHP," and "DOP" is normally reserved for dioctlyl phthalate.

(PVC) resin with a degree of polymerization of 1100, melted in a Banbury mixer, and formed into a sheet with a thickness of 200  $\mu$ m by using a calendering machine. The reinforcing agent was one formed from a methyl methacrylate, butadiene, styrene, and acrylic acid ester polymer, and the stabilizer was a Ba-Zn stabilizer.

[0024] [Table 5]

	PVC	MBS	DOP	Stabilizer
Comparative Example 6	100 parts	10 parts		3 part
Comparative Example 7	100 parts	10 parts	10 parts	3 part
Comparative Example 8	100 parts	10 parts	30 parts	3 part
Comparative Example 9	100 parts	10 parts	50 parts	3 part

[0025] Tensile strength (MPa) and elongation (%) were measured at an elastic stress rate of 200 m/min and temperatures of 23°C and 40°C by using samples that had a length (in the drawing direction) of 80 mm and a width of 10 mm and were drawn from the sheets obtained in Working Examples 1 to 4 and Comparative Examples 1, 2, 4 and 6 to 9. The results are shown in Table 6. In Table 6, the asterisks designate cases in which a sheet could not be formed. [0026]

[Table 6]

	Tensile strength (MPA)		Elongation (%)	
	23°C	40°C	23°C	40°C
Working Example 1	47	15	4	250
Working Example 2	22	11	383	230
Working Example 3	44	20	6	220
Working Example 4	25	12	335	251
Comparative Example 1	44	34	5	4
Comparative Example 2	42	30	4	8
Comparative Example 3	*	*	*	*
Comparative Example 4	44	36	4	6
Comparative Example 5	*	*	*	*
Comparative Example 6	54	41	35	151
Comparative Example 7	44 ,	28	59	225
Comparative Example 8	39	22	225	249
Comparative Example 9	22	11	228	244

[0027] A comparison of Working Examples 1 and 2 and of Comparative Examples 3 and 4 indicates that the tensile strength and elongation in these examples are different. Based on this, it can be concluded that the sheet hardness varies with the amount in which the additive is added. It can also be seen that the sheets of Working Examples 1 to 4 exhibit the same softening behavior as the vinyl chloride resin sheets of Comparative Examples 6 to 9. The sheets of Comparative Examples 1, 2, and 4 did not exhibit any softening behavior. In Comparative Examples 3 and 5, the material was too soft and could not be formed into a sheet.

[0028] High-frequency welding and gravure printing tests were performed using the sheet of Working Example 1, and satisfactory processing results were obtained. It was concluded based on these results that the resin sheet of the present invention can be softened and endowed with properties or behavior traits that are similar to those of a vinyl chloride resin sheet while allowed to retain the characteristics of a noncrystalline polyester resin.

#### [0029]

[Effect of the Invention] The resin for use in sheets and the resin sheet of the present invention exhibit properties or behavior traits that are similar to those of a vinyl chloride resin and sheet, and can be easily handled in the same way as a vinyl chloride resin. The resin and sheet also have excellent fabrication properties, such as being able to be made into sheets by calendering, permitting hardness to be freely adjusted by varying the amount of the introduced additive, exhibiting adequate elongation at low temperatures (23°C to 40°C), and allowing printing, welding, and other operations to be performed without any pretreatment.

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